

Pt/SnO₂ CATALYSTS FOR NO DECOMPOSITION

Ates Akyurtlu, Jale F. Akyurtlu, Wesley Bridges, and
Jillyan Harlan

Grant No: DE-FG26-03NT41911

Hampton University, Department of Chemical Engineering,
Hampton, VA

Presented at the 2005 UCR/HBCU and OMI Contractors Review Conference

WHY Pt/SnO₂

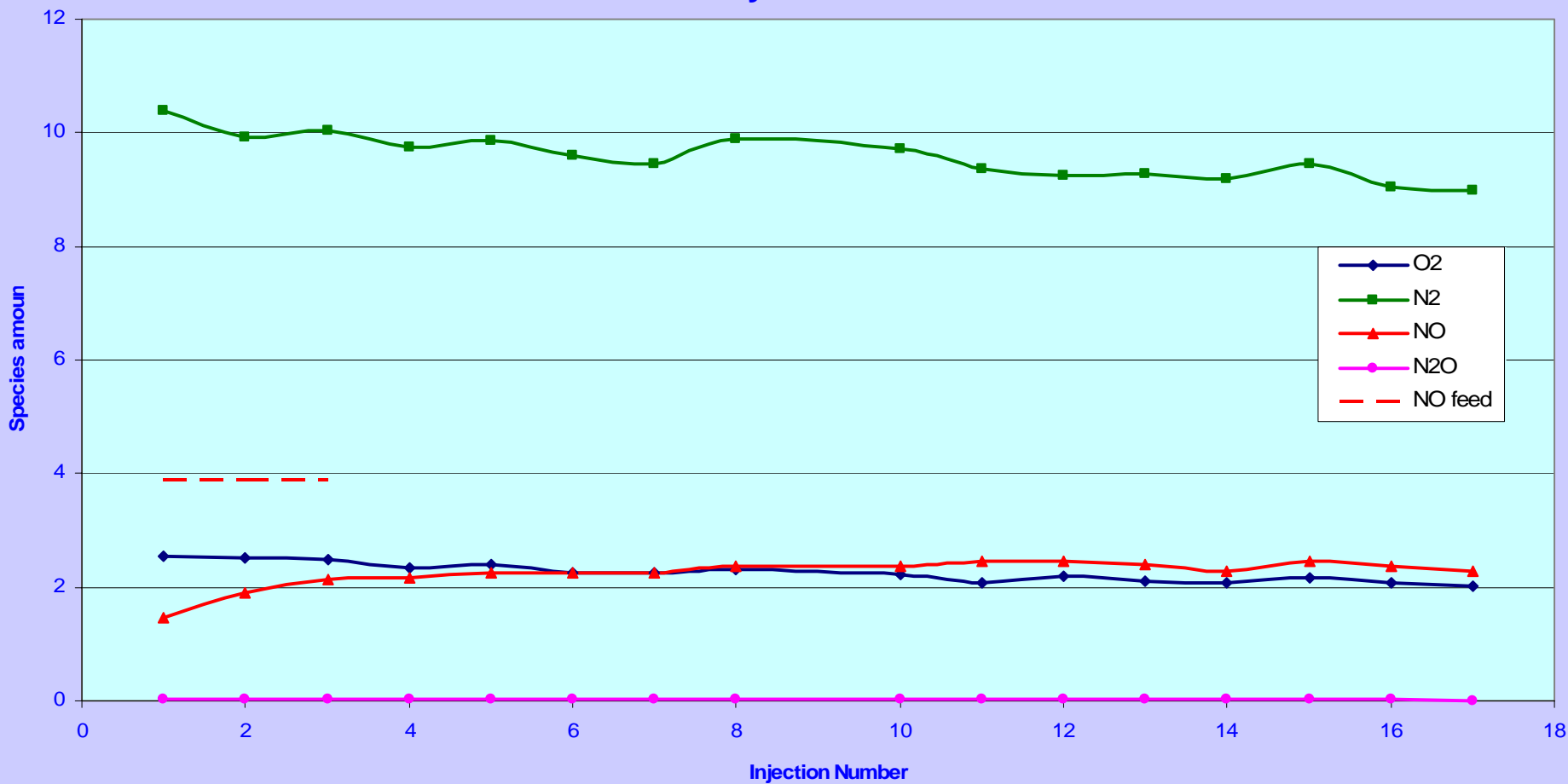
The most promising decomposition catalysts are transition metal-exchanged zeolites, perovskites, and noble metals supported on metal oxides such as alumina, silica, and ceria. The main shortcoming of the noble metal reducible oxide catalysts is that they are prone to deactivation by oxygen.

WHY Pt/SnO₂

Catalysts containing tin oxide show oxygen adsorption behavior that may involve hydroxyl groups attached to the tin oxide. This observation leads one to believe that the Pt/SnO₂ catalysts may have a potential as NO decomposition catalysts in the presence of oxygen.

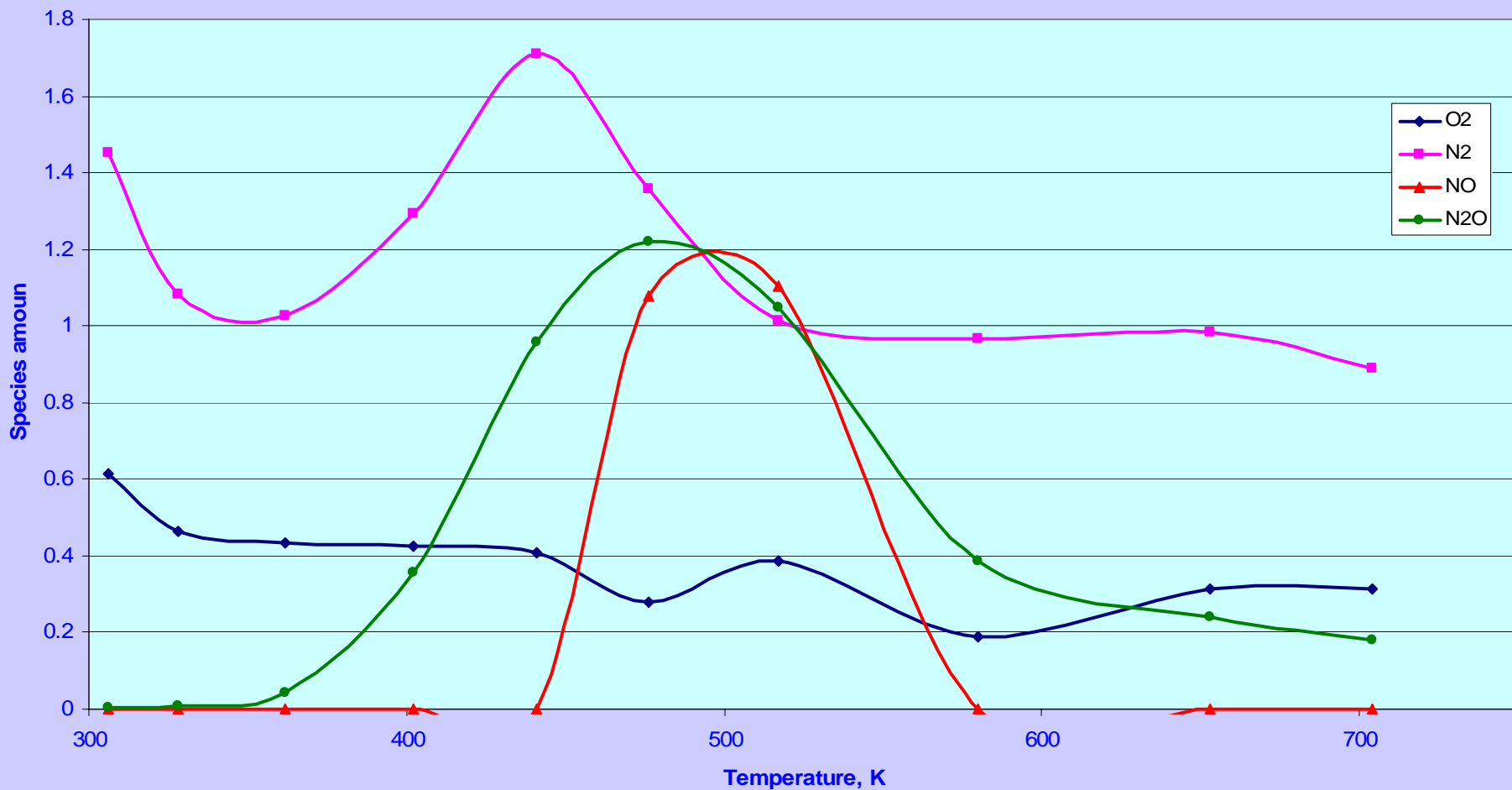
Temperature-Programmed Desorption Experiments

Species evolved during injection of 1-ml doses of 1.94% NO in He on 15% Pt/SnO₂ catalyst at 313 K.



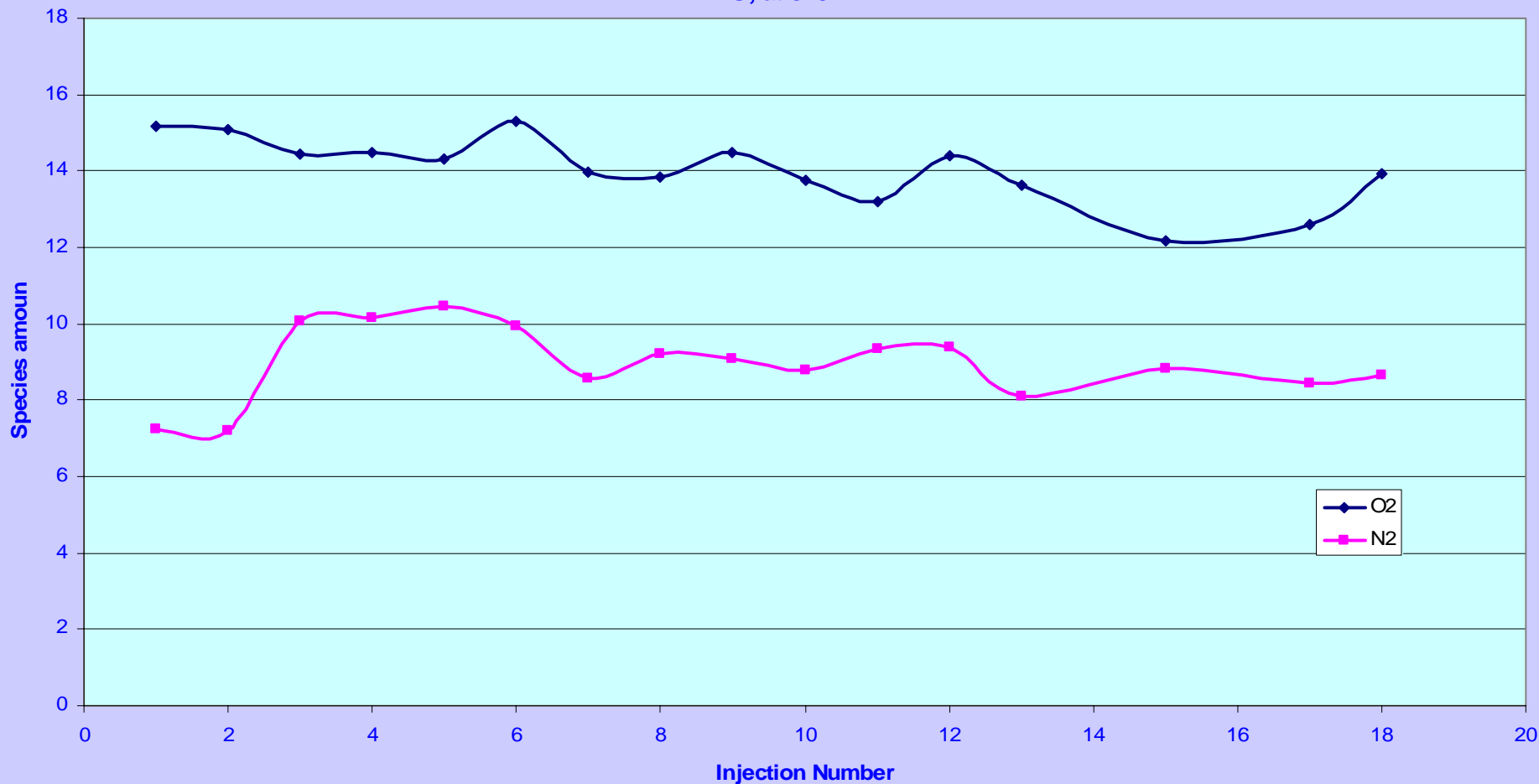
Temperature-Programmed Desorption Experiments

Species evolved during TPD after treatment of 15% Pt/SnO₂ catalyst with NO at 313 K.



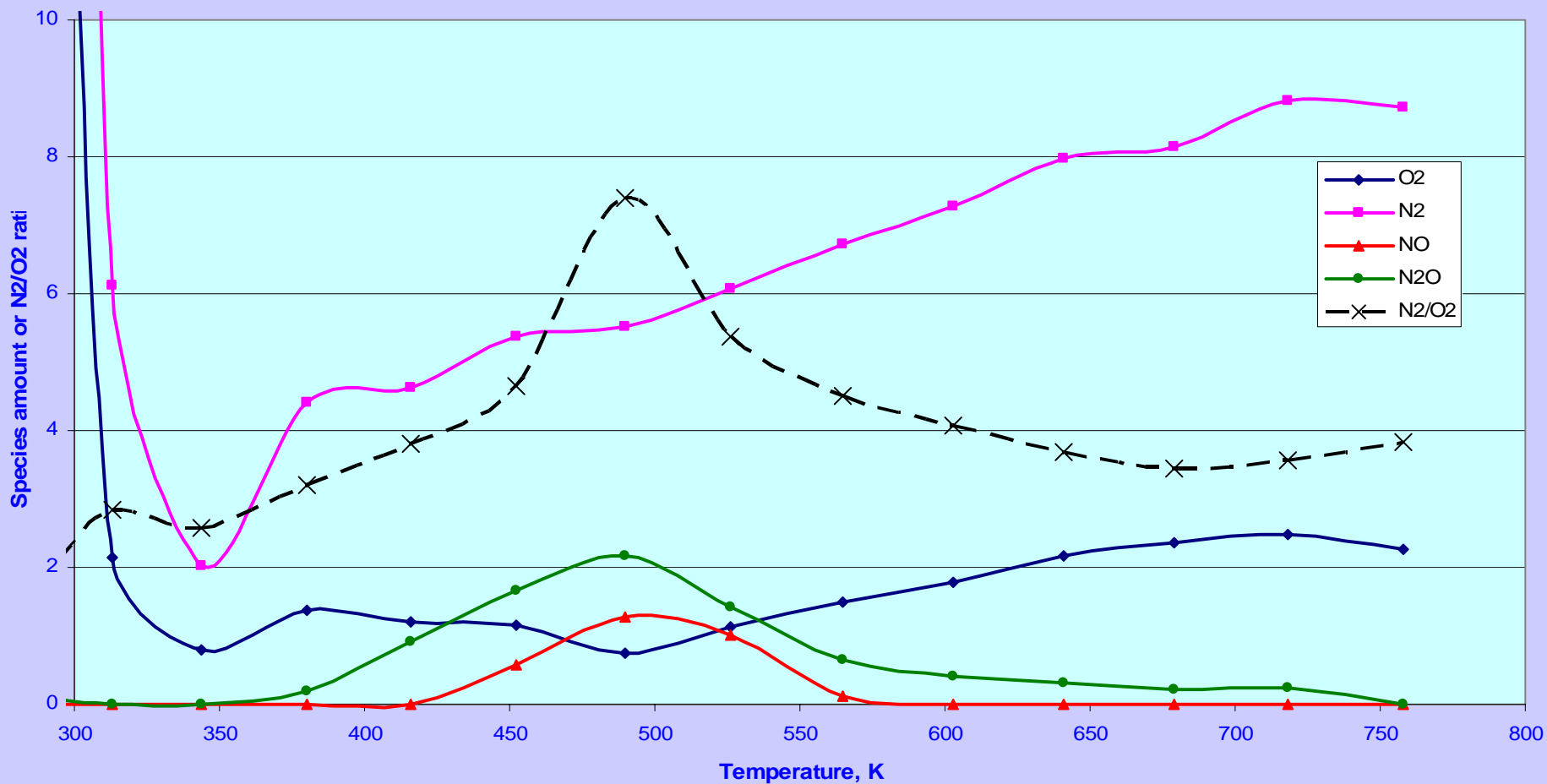
Temperature-Programmed Desorption Experiments

Species evolved during injection of 1-ml doses of 3.6% O₂ in He on 15% Pt/SnO₂ catalyst after treatment with NO, at 313 K.



Temperature-Programmed Desorption Experiments

Species evolved during TPD after the treatment of 15% Pt/SnO₂ catalyst with NO and O₂ subsequently at 313 K.



CONCLUSIONS: TP Desorption

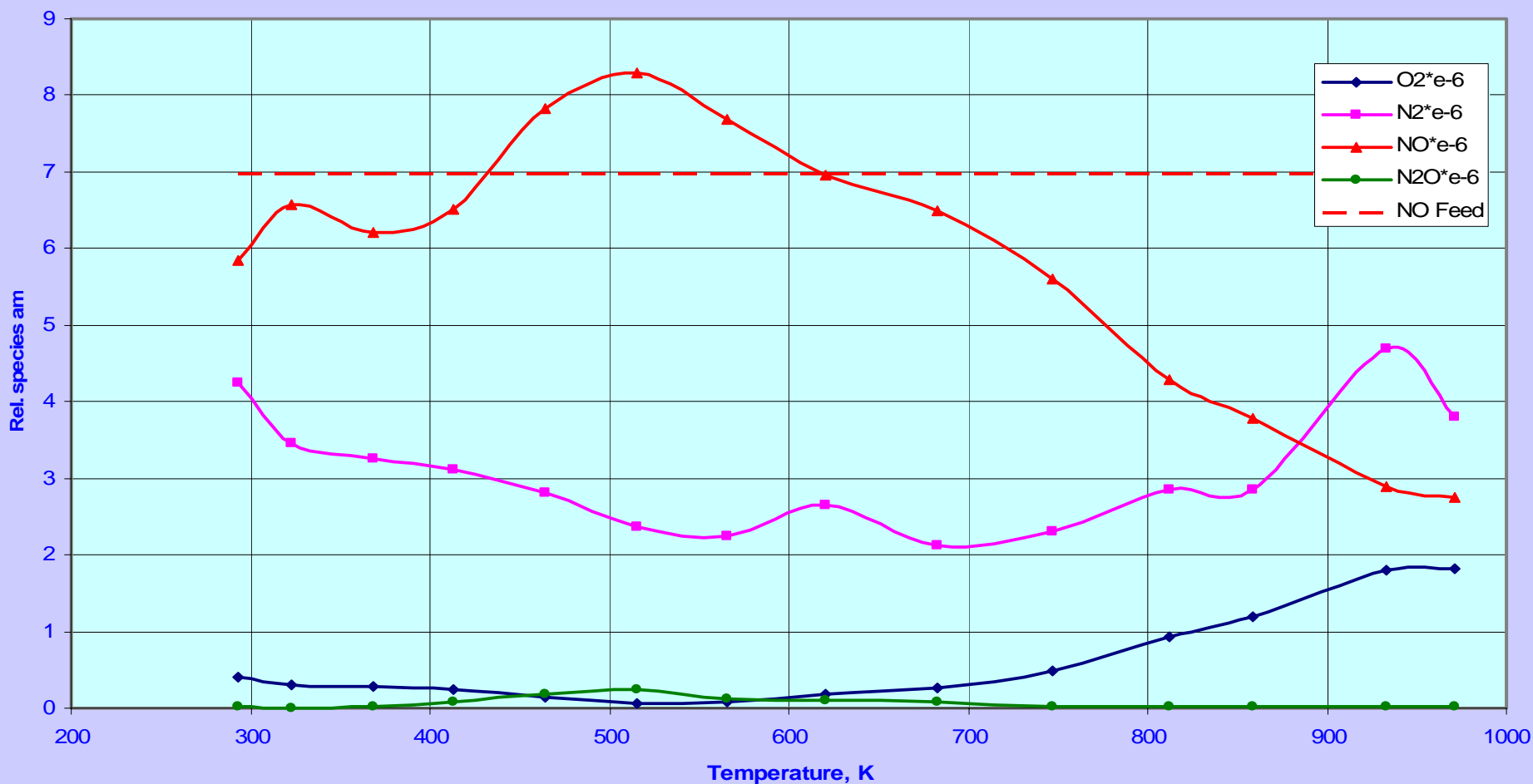
- During pulse chemisorption, about half of NO is either dissociated into N_2 and O_2 or retained on the catalyst at a steady rate. Based on the N_2/O_2 ratio, the latter is deemed a more probable explanation.
- During TPD, the N_2 , N_2O , and NO peaks were separated by about 25 K. There were two O_2 minima, one corresponding to the N_2O peak and the other to the termination of NO peak.
- The observed NO peak could be due to the presence of free surface nitrite or nitrate species, but the presence of N_2O_4 and N_2O_3 is more likely.

CONCLUSIONS: TP Desorption

- The 25 K separation between the N_2 and N_2O peaks suggests that these two species are produced, at least partially, by different surface reaction schemes.
- During the TPD after successive adsorptions of NO and O_2 , the N_2O and NO peaks and the single O_2 minima were all observed around 490 K suggesting that in the presence of excess O_2 , both N_2O and NO were produced by parallel surface reactions.
- Presence of excess oxygen increased the production of N_2O .

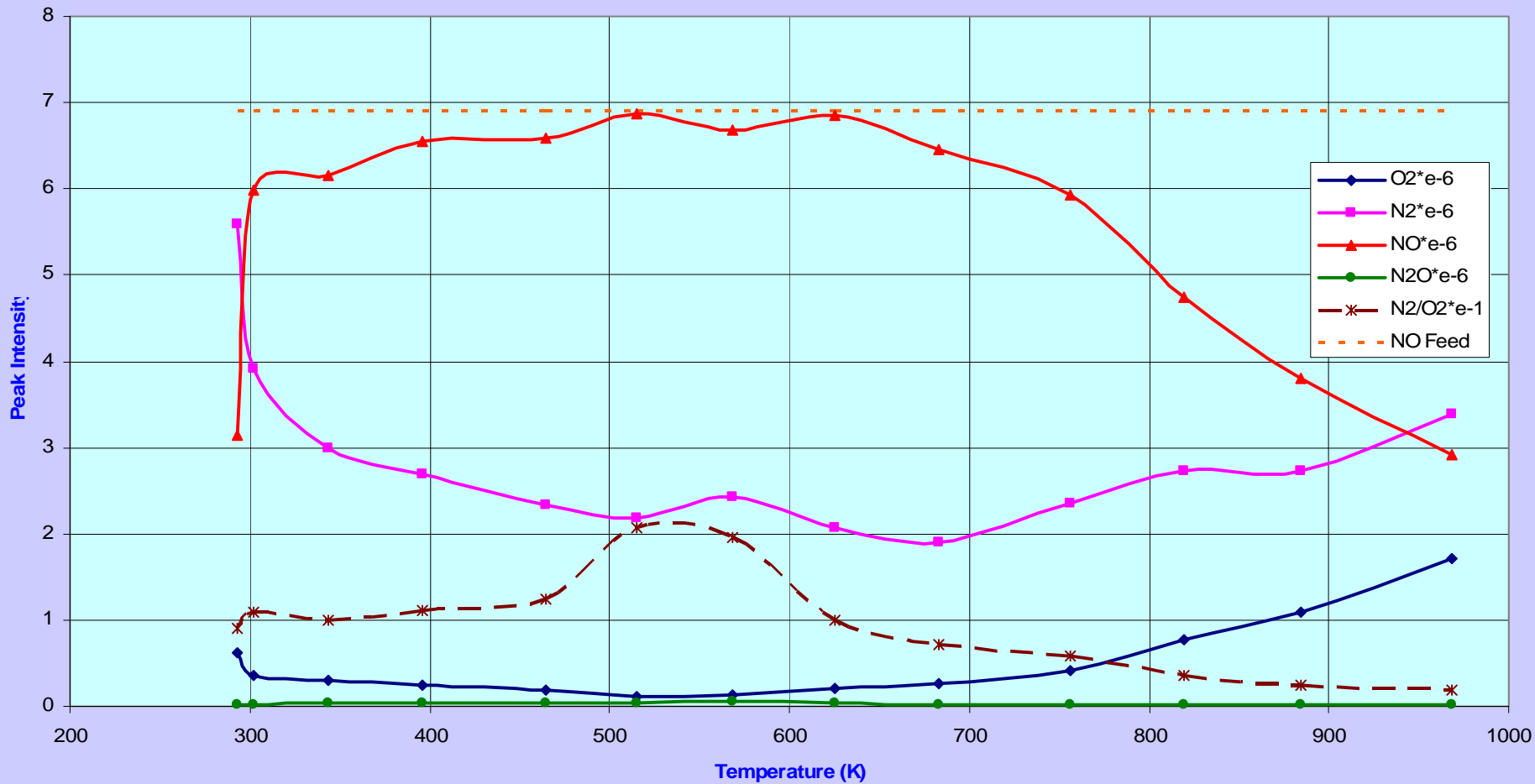
Temperature-Programmed Reaction Experiments

TPReaction of NO on Fresh 15% Pt/SnO₂ Catalyst- Feed Gas: 634 ppm NO in He.



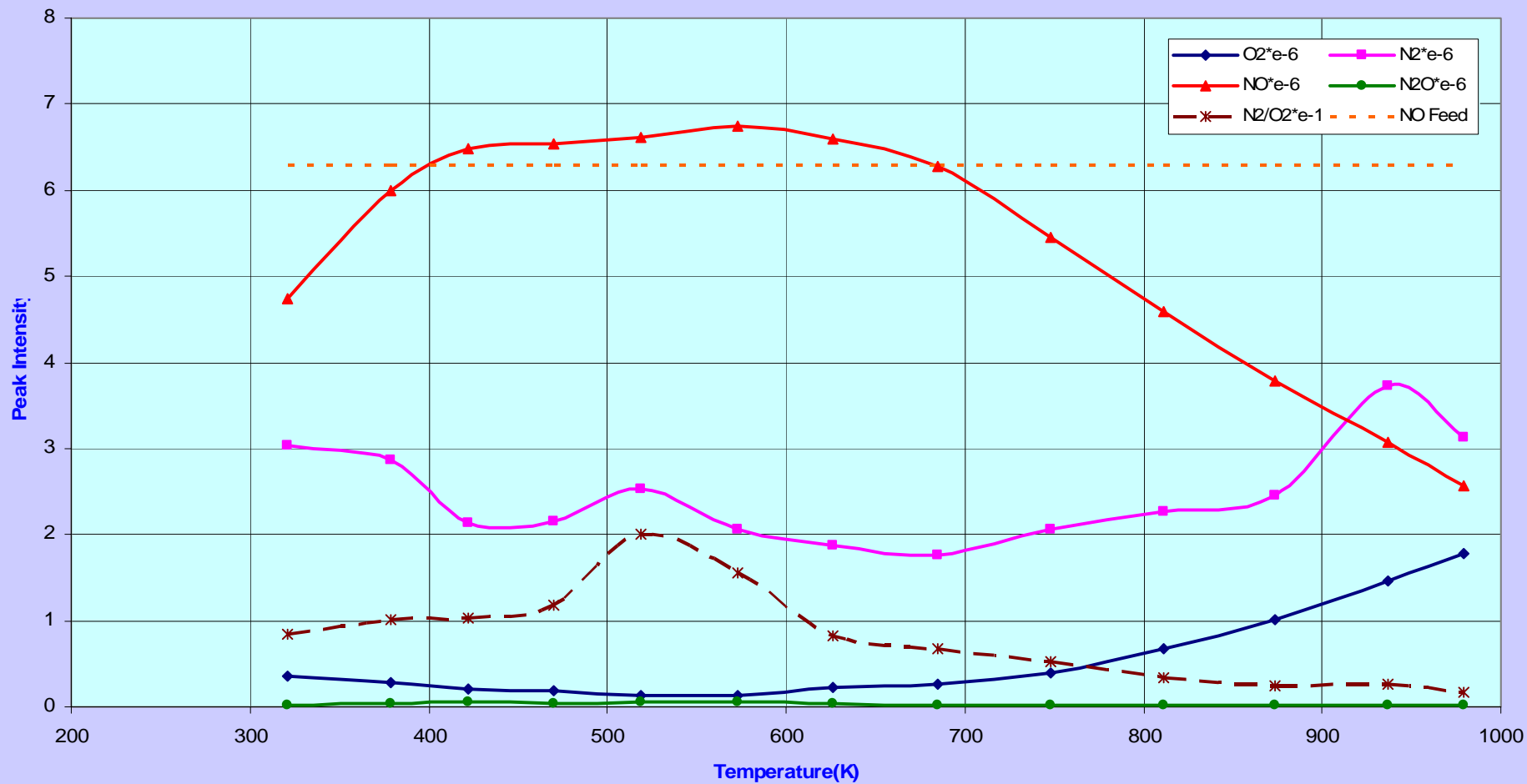
Temperature-Programmed Reaction Experiments

Second TPRx run of NO on 15% Pt/SnO₂ Catalyst.



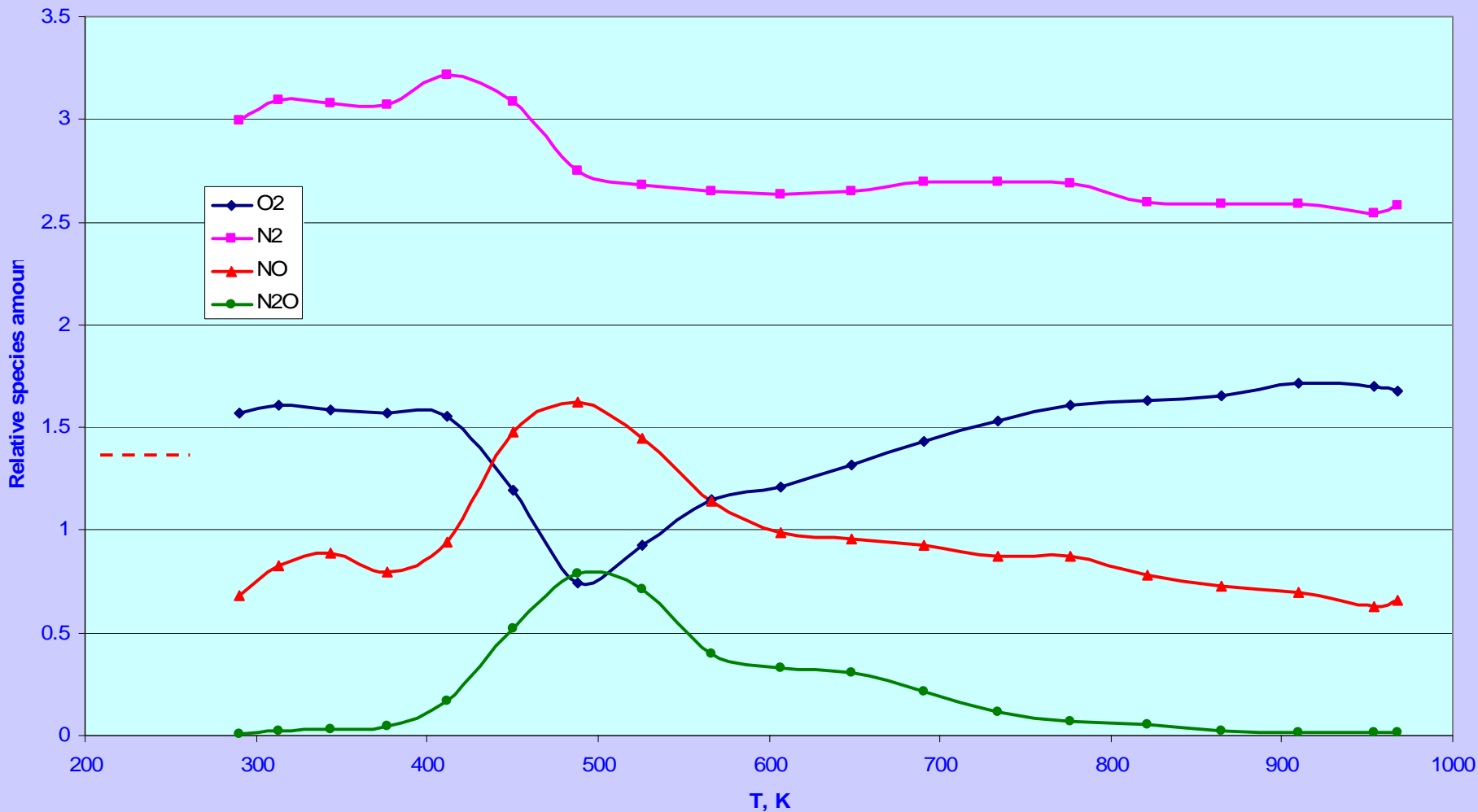
Temperature-Programmed Reaction Experiments

Third TPRx run of NO on 15% Pt/SnO₂ Catalyst.



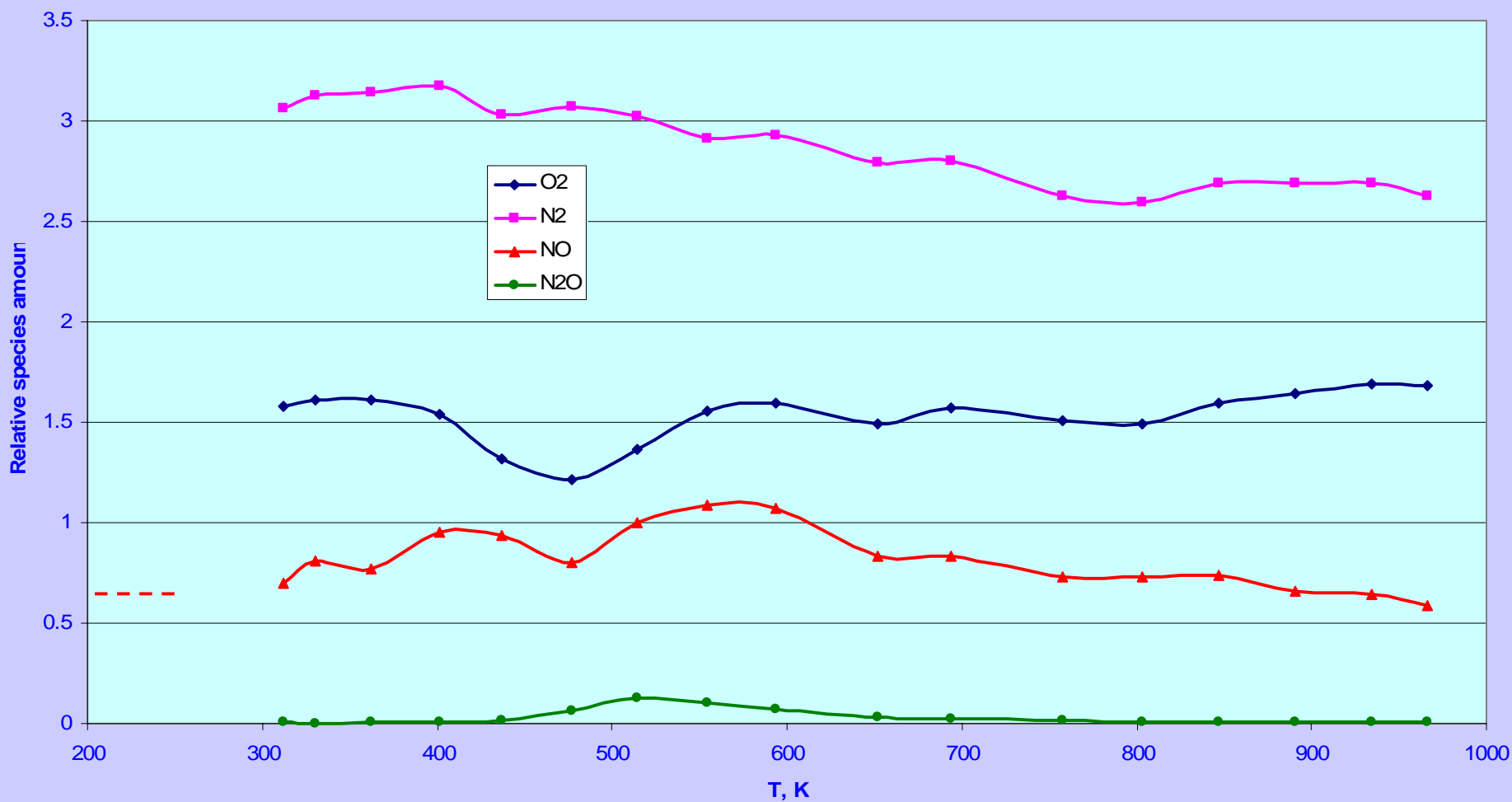
Temperature-Programmed Reaction Experiments

TPRx of NO+O₂ on fresh 15%Pt/SnO₂ catalyst. NO=634 ppm.



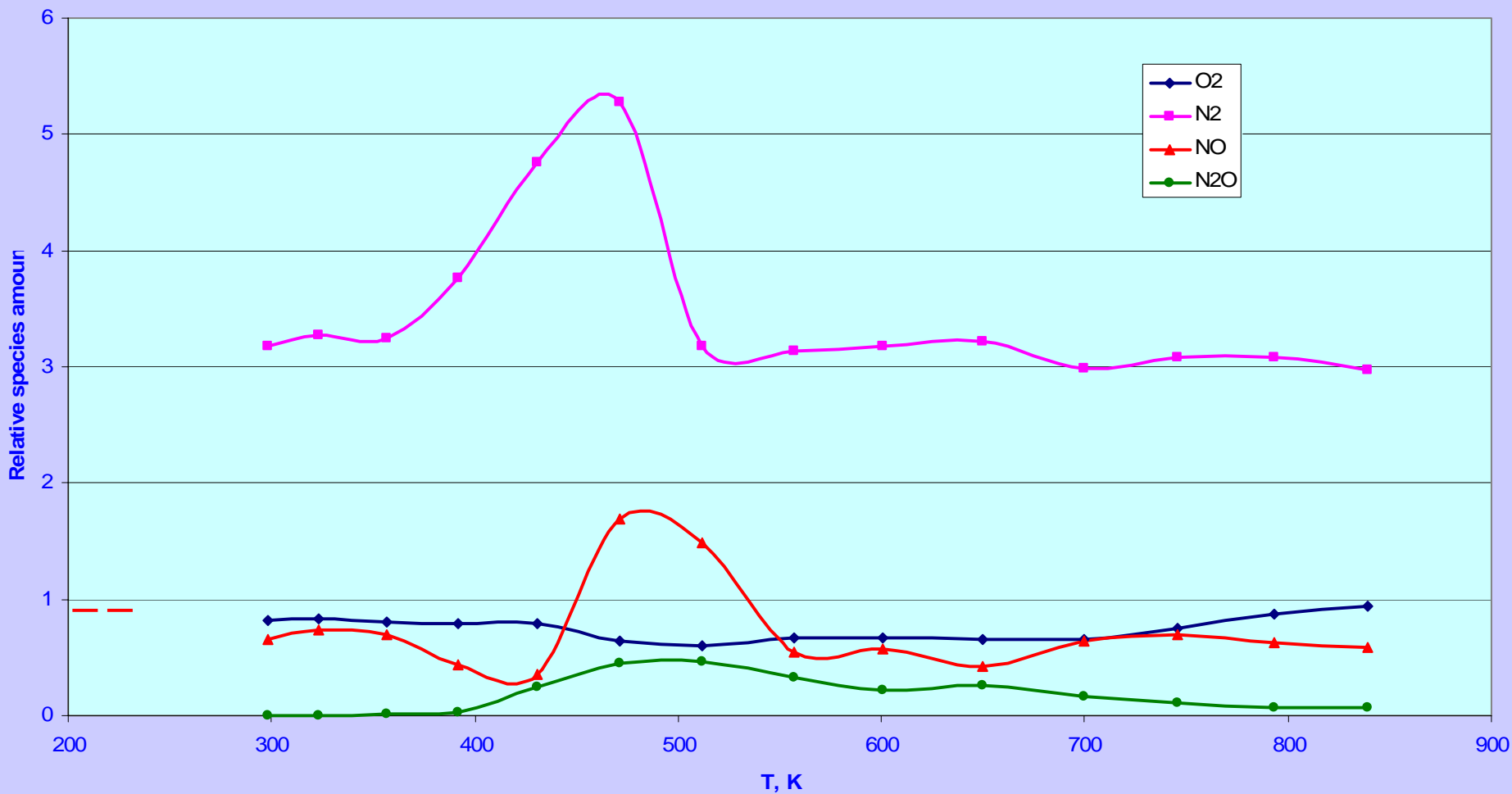
Temperature-Programmed Reaction Experiments

TPRx of NO+O₂ on 15%Pt/SnO₂ catalyst. Second run.



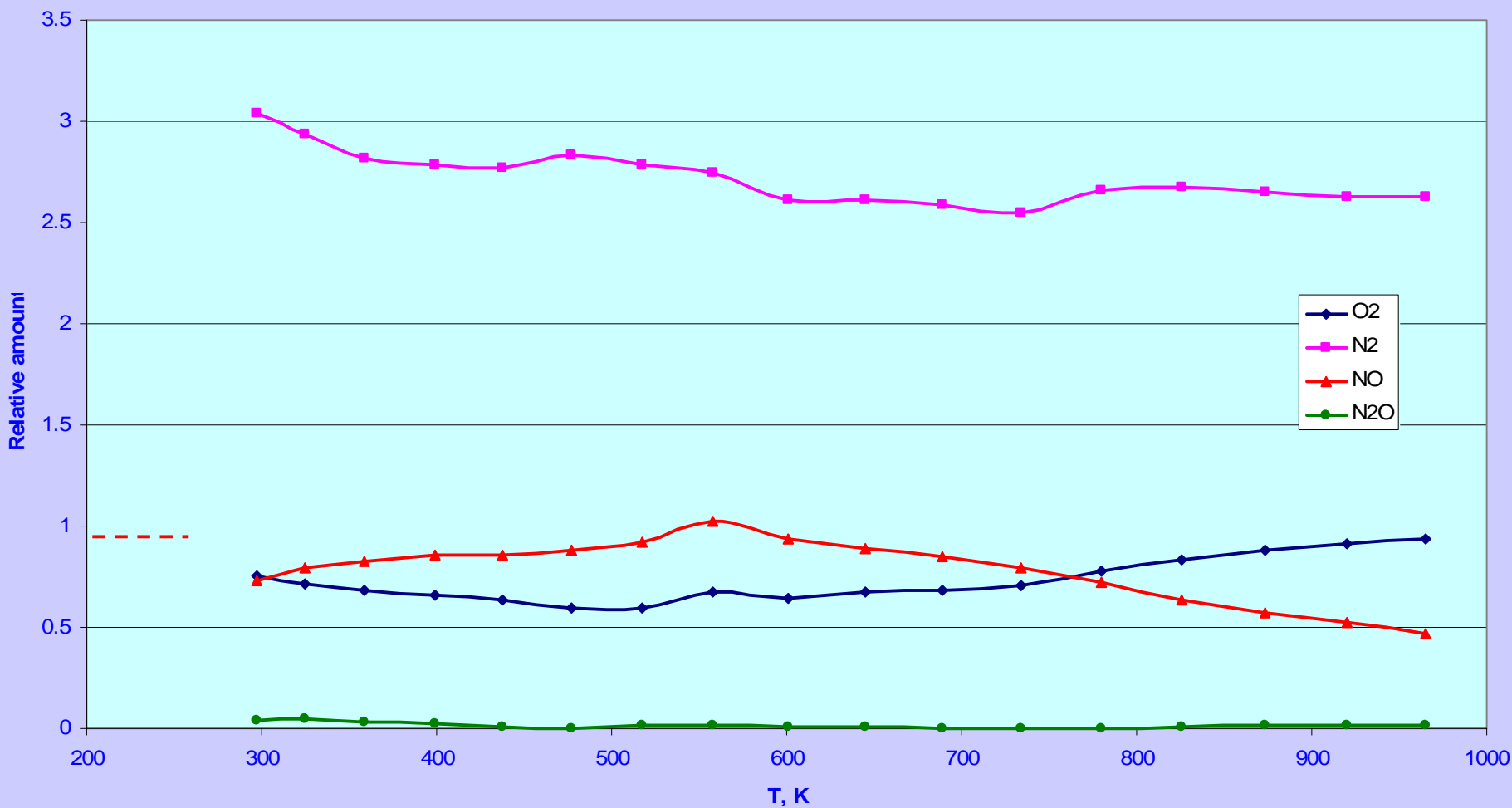
Temperature-Programmed Reaction Experiments

TPRx of NO on fresh 10%Pt/SnO₂ catalyst. NO=634 ppm



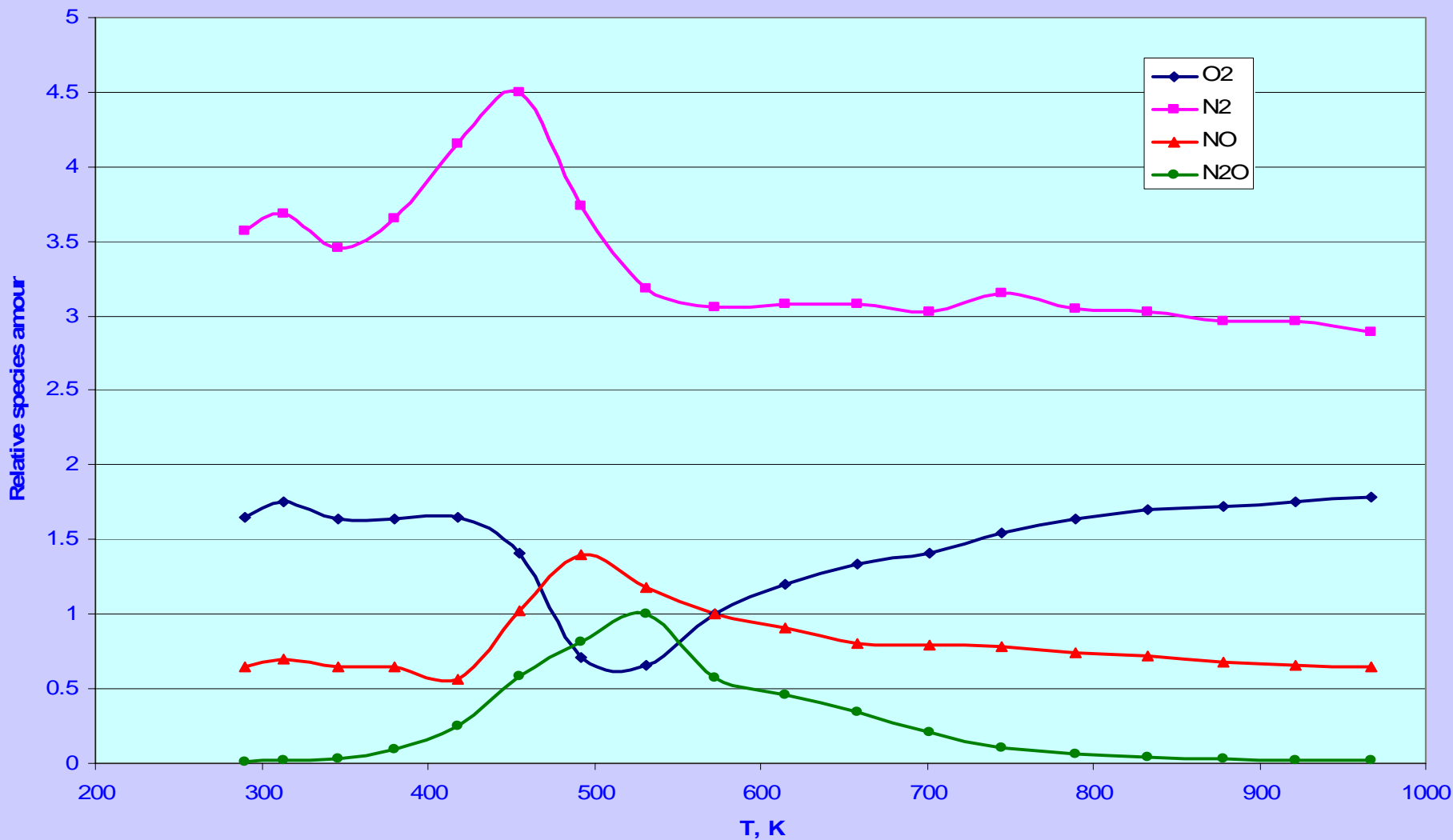
Temperature-Programmed Reaction Experiments

TPRx of NO on 10%Pt/SnO₂ catalyst. Second run. NO=640 ppm



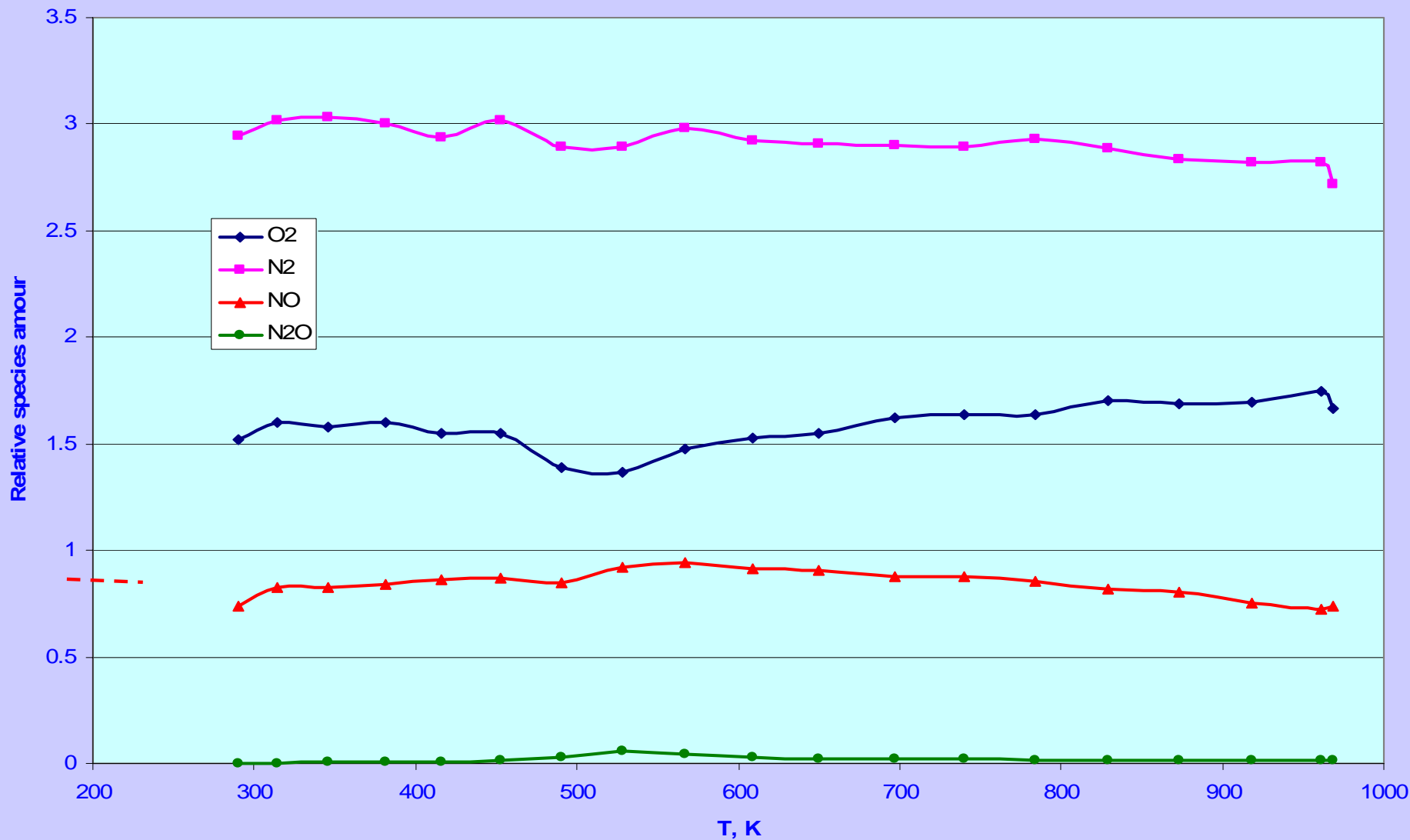
Temperature-Programmed Reaction Experiments

TPRx of NO+O₂ on fresh 10%Pt/SnO₂ catalyst



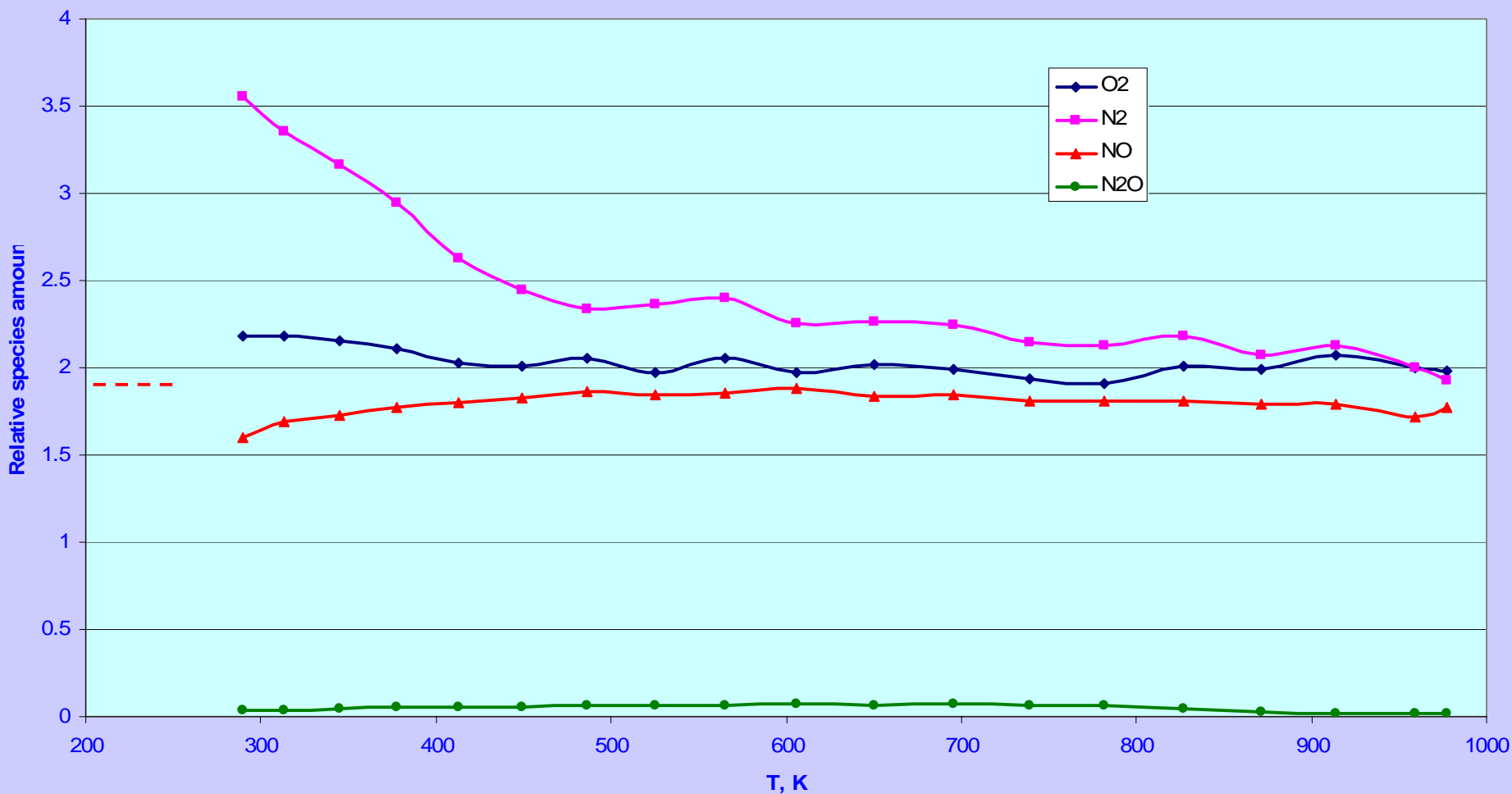
Temperature-Programmed Reaction Experiments

TPRx of NO+O₂ on 10% Pt/SnO₂ catalyst. Second run.



Temperature-Programmed Reaction Experiments

TPRx of NO+O₂ with no catalyst. NO=634 ppm



CONCLUSIONS: TP Reaction of NO

- There was no NO decomposition below 520 K, but some N_2O formation above 370 K, which peaked at 520 K, but no N_2O was present above 750 K. NO desorption started at 520 K and increased with temperature.
- The N_2O formation on the used catalysts was significantly smaller than that on the fresh catalyst.
- The start of NO decomposition is pushed to 625 K during subsequent runs with the same catalyst, but the rate is faster so that the NO concentration in the gas at 900 K is the same for fresh and used catalysts, which corresponds roughly to 50% conversion of NO.

CONCLUSIONS: TP Reaction of $\text{NO} + \text{O}_2$

- The presence of oxygen inhibits the decomposition of NO and promotes the formation of N_2O . On catalysts with 15% Pt, some decomposition activity starts around 425 K on fresh catalyst. During the second run NO decomposition does not start until 575 K. During the second run there was no NO decomposition activity up to about 550 K, but significantly less N_2O formation was observed.
- On fresh catalysts with 10% Pt, there was very little decomposition activity, but at lower temperatures N_2O was formed. During the second run NO dissociation activity increased and N_2O formation decreased.